

Preliminary Amendment  
Attorney Docket No. 052550


**REMARKS**

The above amendments include substitute pages in the specification to reflect the amendment made in the international application and to incorporate the changes made in the same amendment to the claims. Enclosed is an English translation of the PCT Article 34 amendment.

In the event that any fees are due in connection with this paper, please charge our Deposit Account No. 50-2866.

Respectfully submitted,

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# HEAT-RESISTANT COMPOSITE DIAMOND SINTERED PRODUCT AND METHOD FOR PRODUCTION THEREOF

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## TECHNICAL FIELD

The present invention relates to a heat-resistant diamond composite sintered body, and a production method thereof.

## BACKGROUND ART

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Heretofore, there has been known a method for producing a diamond sintered body with a sintering aid, for example, carbonate or metal, such as Co, by use of a conventional ultrahigh-pressure synthesizing apparatus (see the following Patent Publications 1 and 2). There has been known another method for synthesizing a high-hardness diamond sintered body excellent in heat resistance, which comprises performing a sintering treatment under higher pressure/temperature conditions than those in a conventional treatment, using an alkaline-earth metal carbonate as a sintering aid, instead of the metal sintering aid (see the following Non-Patent Publication 1). However, fused carbonate having high viscosity imposes limits on grain size, and thereby these sintered bodies have a relatively large grain size of about 5  $\mu\text{m}$  at minimum.

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There has also been known a sintering method free of inclusion, which comprises enclosing in a metal capsule a diamond powder added with no sintering aid, and applying directly to the capsule an external ultrahigh pressure instantaneously generated primarily from burst of an explosive, or hitting against the capsule a metal plate blown at high speed by an ultrahigh pressure generated from burst of an explosive or the like, so as to compressingly form a shock-synthesized diamond power. However, if this method uses a diamond powder having a grain size of 250 to 500 nm, which is synthesized by a static compression process, any high-hardness diamond sintered body cannot be obtained due to graphitization in a part of the diamond powder.

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As one of measures for this problem, there has been known a method for producing a diamond consisting of ultrafine grains through a shock-compression process using shock-synthesized polycrystalline diamond grains (Patent Publication 3). The Patent Publication 3 discloses Example 4 where shock-synthesized polycrystalline diamond grains having a grain size of 100 to 500 nm are sintered through a shock-compression process at a pressure of 71.8 GPa with a reaction time of several ten  $\mu$ sec. This sintered body has a hardness of 5000 to 6700 Kg/mm<sup>2</sup> (49 to 65.7 GPa), and includes a small amount of graphite. That is, the hardness of this sintered body is low as compared to about 100 GPa in a natural single crystal.

The inventors reported a method for producing a fine-grain diamond sintered body, which comprises adding oxalic acid dihydrate serving as a source of a CO<sub>2</sub>-H<sub>2</sub>O fluid phase into carbonate to prepare a mixed powder, and applying a natural diamond powder having a grading range of zero to 1  $\mu$ m, onto the mixed powder to form a layered structure (see the following Patent Publication 4 and Non-Patent Publications 2 and 3). However, this production method essentially requires a high temperature of 2000°C or more.

The inventors also reported a method similar to the above method, which comprises sintering a finer-grain diamond powder, for example, having a grading range of zero to 0.1  $\mu$ m (see the following Non-Patent Publication 4). In this case, any high-hardness diamond sintered body could not be obtained due to occurrence of abnormal grain growth in diamond.

Recently, an article has been published that discloses a method for synthesizing a diamond sintered body under a pressure of 12 to 25 GPa at a temperature of 2000 to 2500°C without a sintering aid through a direct conversion reaction from graphite to diamond. This article reports that the obtained diamond sintered body has light-transparency (see the following Non-Patent Publication 5).

Parent Publication 1: Japanese Patent Publication No. 52-012126

Parent Publication 2: Japanese Patent Publication No. 04-050270

Parent Publication 3: Japanese Patent Laid-Open Publication No. 02-030668

Parent Publication 4: Japanese Patent Laid-Open Publication No. 2002-187775

10 Non-Patent Publication 1: Diamond and Related Mater., Vol. 5, pp 34-37, Elsevier Science S. A., 1996

Non-Patent Publication 2: Journal of the 41st High Pressure Symposium, p 108, the Japan Society of High Pressure Science and Technology, 2000

15 Non-Patent Publication 3: Proceedings of the 8th NIRIM International Symposium on Advanced Materials, pp 33-34, the National Institute for Research in Inorganic Materials, 2001

Non-Patent Publication 4: Journal of the 42nd High Pressure Symposium, p 89, the Japan Society of High Pressure Science and Technology, 2001

20 Non-Patent Publication 5: T. Irifune et al., "Characterization of polycrystalline diamonds synthesized by direct conversion of graphite using multi anvil apparatus, 6th High Pressure Mineral Physics Seminar, 28 August, 2002, Verbania, Italy

## DISCLOSURE OF INVENTION

There is the need for providing a diamond sintered body usable as a high-performance tool in the field of cutting tools and an ultraprecision machining tool as an alternative to single crystals previously limited largely, and valuable as jewelry. Particularly, in connection of higher cutting speed in oil-drilling bits and particular automobile components, it is desired to achieve enhanced heat resistance in diamond sintered-body tool.

Heretofore, with a sintering aid without distinction of metal or nonmetal, a high-hardness diamond sintered body has been produced through a high-pressure/high-temperature sintering treatment under an ultrahigh pressure condition of 5.5 to 7.7 GPa. In such a diamond sintered

What is claimed is:

1. A heat-resistant diamond composite sintered body prepared by sintering an ultrafine-grain synthetic diamond powder having an average grain size of 200 nm or less, by use of an  
5 ultrahigh-pressure synthesizing apparatus through static compression process without using a sintering aid, said composite sintered body comprising a diamond crystal and a minute amount of non-diamond carbon as a product, and having a Vickers hardness of 85 GPa or more.
2. A method of producing the heat-resistant diamond composite sintered body as defined in  
10 claim 1, comprising:
  - enclosing a synthetic diamond powder having an average grain size of 200 nm or less, in a capsule made of Ta or Mo;
  - placing the capsule in a pressure medium; and
  - heating and pressurizing the capsule under thermodynamically stable conditions
- 15 including a temperature of 2100°C or more and a pressure of 7.7 GPa or more, by use of an ultrahigh-pressure synthesizing apparatus through static compression process.